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The Structure of Soda-Lime-Silicate Glass and Melt by X-ray Diffraction Method

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The structure of soda-lime-silicate($16\text{Na}_2\text{O}\cdot 10\text{CaO}\cdot 74\text{SiO}_2$) glass and melt ($\sim 1500^\circ\text{C}$) was investigated by X-ray diffraction method in combination with molecular dynamics(MD) calculation. The short- and intermediate range structure of the glass was investigated as a function of temperature by using MD simulation. The experimental results clearly show that there is no distinct difference in structure between the present glass and melt. We also found from MD calculation that the linkage of Si-O network made of SiO_4 tetrahedra shows similarities between glass and melt. It is also found that the first sharp diffraction peak(FSDP) associated with intermediate range structure is contributed from the correlation between Na^+ (or Ca^{2+}) and SiO_4 network.

Keywords : Silicate Glass / Melt/ Structure / X-ray diffraction / Molecular Dynamics Calculation

As well known, many commercial glasses are produced through melting process. Although enormous efforts have been made to investigate their physical and chemical properties in a melting state, such as viscosity, density and redox reaction etc., there are few works on the structure of oxide melts. On the other hand, MD calculation is regarded as one of the most powerful techniques to get insight into materials¹. By a combination of high temperature X-ray radial distribution function(RDF) analysis and MD calculation, it is expected that we can get not only information about the average structure of specific elements but also structural parameters of short and medium range orders in glasses and melts.

In the present study, the technologically important soda-lime-silicate glass is chosen to investigate the local structure of glass and melts. The difference in structure between glass and melt is discussed with the result of

MD calculation in order to explain the temperature dependence of macroscopic properties.

Figure 1 shows the interference functions of glass and melt obtained from X-ray diffraction together with the MD-derived ones. It is seen that the interference functions from MD calculation are in fairly well agreement with the experimental ones indicating that the MD simulation reproduces the characteristics of the real structures. It is also seen from Figure 1 that there are two peaks in the low- Q region at 15 and 21nm^{-1} which are hereafter referred to Q_1 and Q_2 , respectively. It is clear that the Q_1 peak disappears at high temperature, while the Q_2 becomes more pronounced in a melting state. Our MD calculation also reproduced this result. By comparison between the total $Q_i(Q)$ and pair $Q_i(Q)_{ij}$, it is obvious that the peaks at $\sim 15\text{nm}^{-1}$ and $\sim 21\text{nm}^{-1}$ are assigned to Si-O(Si-Si) and Na(Ca)-Si correlations, respectively(see Figure 2).

SOLID STATE CHEMISTRY - Amorphous Materials -

Scope of Research

Inorganic amorphous materials with various functions are the targets of research in this laboratory. (1) To obtain a clear view of glass materials and the bases for designing functional glasses, we investigate the structure of glasses using X-ray and neutron diffraction analysis, high resolution MAS-NMR, and ab initio MO calculation. (2) To develop materials with high optical nonlinearity, we search heavy metal oxide-based glasses and transition metal oxide thin films, and evaluate the nonlinear optical properties by Z-scan methods. (3) Photosensitivity of glasses for optical fibers and waveguides is investigated to design efficient fiber gratings and optical nonlinear materials. (4) Using sol-gel method, synthesis and microstructure control are carried out on various functional oxide thin films.



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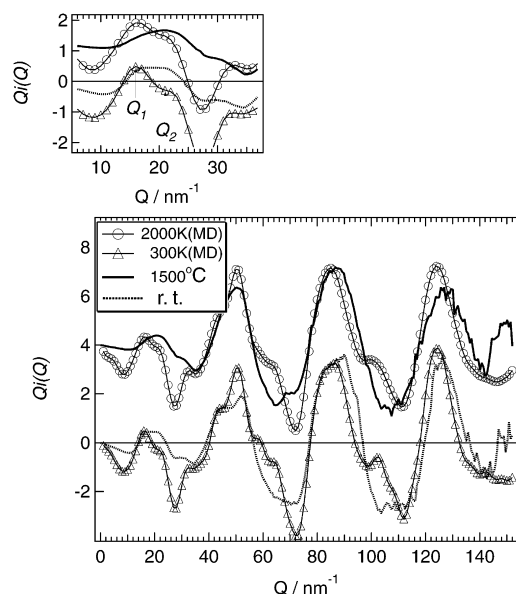


Figure 1. The interference functions of $16\text{Na}_2\text{O} \cdot 10\text{CaO} \cdot 74\text{SiO}_2$ glass and melt.

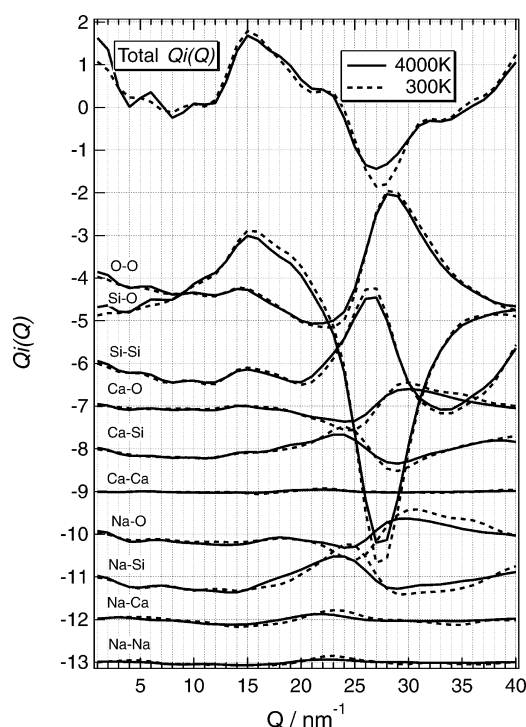


Figure 2. MD results for partial and total interference function of $16\text{Na}_2\text{O} \cdot 10\text{CaO} \cdot 74\text{SiO}_2$ glass and melt.

Namely, the former corresponds to network and the latter to correlations between cation and network of glass. The previous result from MD calculation of pure SiO_2 also shows the FSDP at $\sim 15\text{nm}^{-1}$ which is a contribution of Si-Si, Si-O and O-O pairs^{2,3}. In our MD calculation, the peak at $\sim 21\text{nm}^{-1}$ is shifted to low Q side on increasing temperature. It indicates the expansion of Na(Ca)-Si correla-

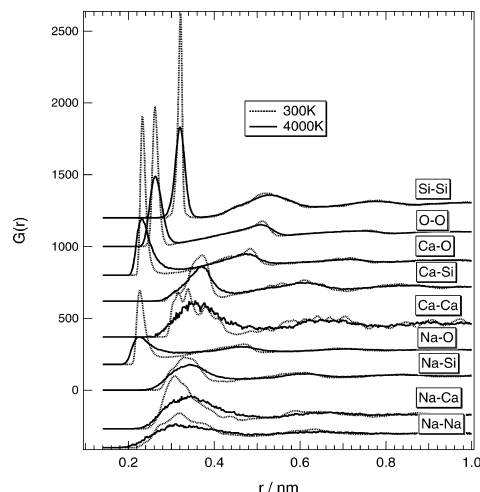


Figure 3. Pair distribution functions of $16\text{Na}_2\text{O} \cdot 10\text{CaO} \cdot 74\text{SiO}_2$ glass and melt derived from MD calculation.

tion due to thermal motion. Now we can draw a picture that the distance between the modifier cations and the network former in glass tends to expand with increasing temperature.

It is surprising that almost no change of Si-O and Si-Si atomic distances was observed between the RDF curves of glass and melt. However, we have observed that the atomic distances of Na-Ca and Ca-Ca were increased by 0.031nm and 0.019nm, respectively, with increasing temperature (see Figure 3). This value is longer than expansion of Si-O at 1000K (0.002nm)⁴. The distinct change of distribution of Na-O and Ca-O correlations that are difficult to be distinguished in experimental $G(r)$ with increasing temperature was also observed. This allows us to suggest that the temperature dependence of density cannot be simply explained by the change of local structure of network former and instead the thermal motion of Na^+ and Ca^{2+} ions mainly contributes to the expansion of glass at high temperatures.

In the present study, we have investigated the structure of soda-lime-silicate ($16\text{Na}_2\text{O} \cdot 10\text{CaO} \cdot 74\text{SiO}_2$) glass and melt ($\sim 1500^\circ\text{C}$) by X-ray diffraction method in combination with MD calculation. The experimental results clearly show that there is no distinct difference in structure between the present glass and melt, which was also supported by MD calculation. The decrease in density with increasing temperature is due to expansion of atomic distance between Na^+ and Ca^{2+} (Na^+). It is concluded that the increase of correlation length between cations and network with increasing temperature is also responsible for the decrease of density.

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